Atomic Ion Frequency Standards

WAYNE M. ITANO

Invited Paper

A new class of atomic frequency standard, based on ions trapped by electromagnetic fields, is under development. Such standards have the potential of achieving higher frequency accuracy than currently available standards. They are also capable of very good frequency stability. The history and status of trapped-ion frequency standards are reviewed. Prospects for future standards are discussed.

I. Introduction

Frequency standards based on trapped ions are still in a very early stage of development. Nevertheless, they appear to have some fundamental advantages over the more established types of frequency standards. Several reviews on ion traps and their application to frequency standards have already appeared [1]-[4].

In a trapped-ion frequency standard, the frequency of an oscillator is servoed to a resonance which corresponds to a transition between two energy levels of an atomic ion. The ions are suspended in space by a combination of electric and magnetic fields. In a conventional rubidium cell, the atoms are surrounded by a buffer gas having a pressure of about 10³ Pa (approximately 10 torr). In an ion trap, the ions are held either in a vacuum or in a low-pressure buffer gas (less than 10⁻³ Pa). In an atomic beam, the atoms also move through a vacuum, without collisions. However, the time available for interaction with the electromagnetic field is limited to their flight time through the apparatus, usually about 10 ms or less. Trapped ions can be observed for much longer periods.

Several types of trapped-ion frequency standards are currently under development. The ¹⁹⁹Hg⁺ microwave frequency standard is the best developed [5]–[9]. Another frequency standard is based on a 303-MHz transition in ⁹Be⁺ [10]–[12]. Other microwave frequency standards are based on ¹³⁷Ba⁺ [13] or ¹⁷¹Yb⁺ [14]. Optical frequency standards, based on narrow linewidth transitions in single trapped ions are being investigated [15]–[17]. They may

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The author is with the Time and Frequency Division, National Institute of Standards and Technology, Boulder, CO 80303.

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eventually have accuracies several orders of magnitude better than any present-day standards [18], [19].

II. ION TRAPS

Two basic types of ion traps have been used in frequency standards. These are the Penning trap, also called the electromagnetic trap, and the Paul trap, also called the electrodynamic or radiofrequency (RF) trap [1], [3].

A. The Penning Trap

The Penning trap uses static electric and magnetic fields. The electrostatic potential is

$$\phi(x, y, z) = A(x^2 + y^2 - 2z^2). \tag{1}$$

A typical electrode configuration used to create such a potential is shown in Fig. 1. The two endcap electrodes are held at the same potential relative to the ring electrode. The sign of A is such as to generate an electric field that forces the ion back toward the center if it is displaced in either direction along the z axis of the trap. However, if the ion is displaced radially (that is, in the xy plane) it is subjected to an electric force that forces it away from the center. Superimposing a sufficiently strong magnetic field confines the ions in all dimensions. A single ion undergoes simple harmonic motion along the z axis and a superposition of two circular motions in the xy plane. The higher-frequency motion is called the cyclotron motion; the lower-frequency motion is called the magnetron motion. The magnetron motion is a circular $\vec{E} \times \vec{B}$ drift about the trap axis. Motion along the z axis is stable, because work has to be done to increase z^2 . On the other hand, if the orbit of an ion is displaced radially outward, its potential energy decreases. Hence, energy conservation does not prevent the ions from being lost from the trap. However, conservation of L_z , the z component of the canonical angular momentum of the ions, leads to radial confinement [20]. In a real trap, L_z is only approximately conserved, because of collisions with neutral molecules and because of deviations of the trap electric and magnetic fields from cylindrical symmetry.

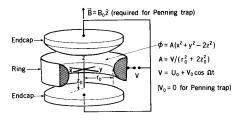


Fig. 1. Standard electrode configuration of a Penning trap or a Paul trap.

B. The Paul Trap

A commonly used form of Paul trap, the RF quadrupole trap, uses an electrode configuration like the Penning trap (Fig. 1). Unlike a Penning trap, the electric potential applied between the electrodes oscillates at a high frequency. The parameter A in (1) has the form

$$A = \frac{U_0 + V_0 \cos \Omega t}{r_0^2 + 2z_0^2}. (2)$$

Here, U_0 and V_0 are the static and RF potentials, and r_0 and z_0 are lengths that depend on the electrode geometry. Dynamic trapping is possible for some range of values of U_0 , V_0 , and Ω . With such values, the time-averaged force (the force averaged over a period of the oscillation) confines an ion in all dimensions. The static part of the electric potential can be adjusted to vary the ratio of the radial and axial restoring forces. The trajectory of an ion is a superposition of a driven motion at frequency Ω and a low-frequency motion, due to the time-averaged force. The driven motion is called the micromotion and the low-frequency motion is called the secular motion.

Other types of RF trap may be useful in frequency standards. One is the linear RF trap. A schematic drawing is shown in Fig. 2. An RF potential is applied between the rods. The phase of the potential at each rod differs by 180° from that of the two that are nearest to it. This creates a time-averaged force which attracts an ion to the central axis. An electrostatic potential applied to the electrodes at the ends prevents the ions from escaping along the axis. In such a trap the RF fields approach 0 along a line rather than at only a point. In this trap the kinetic energy of the micromotion is less than in an RF quadrupole trap with the same number of ions and the same radial restoring force. Such a trap has recently been constructed for a frequency standard by Prestage et al. [21]. A "racetrack" trap can be made by connecting the ends of the rods into rings [22]. Recently, ions have been laser-cooled in a racetrack trap [23].

III. ACCURACY AND STABILITY

Accuracy and stability are distinct properties of frequency standards. Accuracy refers to the absolute reproducibility of the frequency. In practice, it might be defined in terms of the frequency differences between independently constructed and operated standards of the same type. Stability refers to

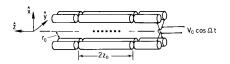


Fig. 2. Electrode configuration of a linear RF trap. Ions are trapped along the central (z) axis.

the uniformity of the average frequency from one interval of time to the next.

A. Accuracy

The accuracy of a trapped-ion frequency standard depends on factors, such as collisions with background gas, perturbations due to external fields, and Doppler shifts, which can shift the resonance frequency of the ions.

In some cases, a buffer gas is deliberately introduced in order to cool the ions, and this leads to a frequency shift [24]. In the case of a frequency standard based on $^9\mathrm{Be^+}$ ions, variations in the background pressure of about 10^{-8} Pa $(10^{-10}$ torr) caused small but observable frequency shifts [12].

The shifts of hyperfine transition frequencies due to the electric fields present in a trap are small enough that they are not a serious problem [25]. This is because both levels are shifted by almost the same amount. Electric fields are more of a problem for frequency standards based on very narrow optical transitions. The rms electric field due to thermal (blackbody) radiation is about 8 V/cm at room temperature. This can cause a shift of an optical transition frequency of about one part in 10¹⁶ from the frequency at 0 K [26]. Also, the energy of a state having an electric quadrupole moment could be shifted by the gradient of the electric field. Such a gradient could be caused by neighboring ions or by static charges on the trap electrodes [18].

The levels involved in the transition must be carefully chosen to avoid shifts due to fluctuating magnetic fields. These are transitions for which $\partial \nu/\partial B$, the first derivative of the frequency ν with respect to the magnetic field B, approaches zero. For example, a transition between two $m_F=0$ sublevels (m_F is the quantum number of the z component of the total angular momentum F) has this property at zero magnetic field. Penning traps are normally operated with magnetic fields of about 1 T. Fortunately, it is possible to find transitions in some ions for which $\partial \nu/\partial B$ approaches zero at a value of B which is high enough to operate a Penning trap.

Cooling the ions is extremely important for reducing Doppler shifts of the resonances. Confinement of an atom to a region smaller than the resonance-radiation wavelength leads to a suppression of the first-order (linear in velocity) Doppler shift. This effect is called Dicke narrowing [27]. If an ion in a three-dimensional potential well is cold enough, its motion can be restricted to less than an optical wavelength, so Dicke narrowing occurs even for an

optical transition. Cooling directly reduces the second-order (quadratic in velocity) Doppler shift.

B. Stability

The stability of a frequency standard is usually described by the sample variance of two successive measurements of the average frequency deviation [28]. This quantity is commonly called the Allan variance $\sigma_y^2(\tau)$, where τ is the measurement time. The square root of the Allan variance typically has the form

$$\sigma_y(\tau) \approx \frac{K}{Q(S/N)} \propto \frac{1}{\sqrt{\tau}}$$
 (3)

where K is a dimensionless constant of order 1, Q is the resonance frequency divided by the linewidth, and S/N is the signal-to-noise ratio for a measurement time τ . In deriving the last part of (3), it was assumed that S/N is proportional to $\tau^{1/2}$. For all frequency standards, $\sigma_y(\tau)$ eventually stops decreasing as τ increases and may even start to rise.

The fundamental limitation on S/N is the statistical fluctuation in the number of ions that make a transition, when subjected to electromagnetic radiation near the resonance frequency. If the frequency is such that half the ions, on the average, make a transition, the fluctuation of the number is $N_i^{1/2}/2$, where N_i is the total number of ions [26].

The Q can be increased by increasing the time taken to drive the transition, since $\Delta \nu$ is inversely proportional to this time. The time cannot be made much longer than the natural lifetime of the upper state, but for microwave transitions, this is not much of a limitation. The other way to increase Q is to increase ν . Hence, there is interest in using narrow optical transitions in frequency standards.

C. Accuracy-Stability Trade-Offs

For trapped-ion frequency standards, there is usually a trade-off between N_i , which limits the stability, and the second-order Doppler shift, which limits the accuracy. In a Penning trap, increasing N_i increases the average $\vec{E} \times \vec{B}$ drift velocity of the ions, since the space charge increases the radial electric fields. In a Paul trap, increasing N_i increases the average velocity due to the micromotion, since the space charge forces the ions away from the center of the trap.

IV. COOLING METHODS

Trapped ions can easily gain several electron volts of kinetic energy, or temperatures of thousands of kelvins, from electric fields in the trap. Since the ions are well isolated thermally from their environment, they do not quickly cool to room temperature. On the other hand, this thermal isolation makes it possible to cool the ions to less than 1 K in a room temperature apparatus, using a weak process like laser cooling.

A. Collisional Cooling

Collisional cooling with neutral gas molecules is a simple way of cooling ions in a Paul trap [29]. Cutler *et al.* [24], [30] have collisionally cooled ¹⁹⁹Hg⁺ ions in a Paul trap with helium gas. The secular motion was cooled to near room temperature, but the micromotion was hotter.

Collisional cooling with neutral atoms is not feasible for ions in a Penning trap. This is so because there is no restoring force in the radial direction. Collisions would quickly drive the ions out of the trap.

B. Laser Cooling

Laser cooling is a very effective method for cooling certain kinds of ions to very low temperatures [31]-[33]. The basic idea is to irradiate the ions with light having a frequency slightly lower than that of a strong resonance line of the ion. Ions moving toward the source of the light absorb and reradiate photons at a high rate, because the Doppler shift brings the light closer to resonance. The ions lose energy, since they absorb the momentum of the photons. When the ions move away from the source of light, the Doppler shift is away from resonance, and photons are scattered at a low rate. The velocity is damped, on the average. The minimum temperature T that can be obtained in this manner is given by $k_BT \approx \hbar \gamma/2$, where k_B is Boltzmann's constant, \hbar is Planck's constant divided by 2π , and γ is the radiative decay rate of the upper state. For typical cases, T is about 1 mK. This kind of laser cooling is called Doppler cooling, to distinguish it from other kinds of laser cooling [33].

Unfortunately, the nearly resonant light field perturbs the transition frequencies of the ion. One way of dealing with this problem is to turn off the light used for cooling for short periods. Another way is to simultaneously trap two kinds of ions. One kind of ion is continuously laser-cooled. It cools the other kind of ion by long range Coulomb collisions. The cooling radiation for one kind of ion does not perturb the resonance frequencies of the other kind very much. This cooling method, called sympathetic laser cooling, was studied by Larson *et al.* [34].

V. RADIOFREQUENCY AND MICROWAVE FREQUENCY STANDARDS

In 1966, the $\Delta F=\pm 1$ ground-state hyperfine transition of $^3\mathrm{He^+}$ was observed by Fortson *et al.* [35]. The method of detecting the resonance was based on collisions with Cs atoms whose electronic spins had been oriented by optical pumping with circularly polarized light. Resonances as narrow as 10 Hz were observed on a 8.666-GHz transition. This corresponded to a Q of almost 10^9 , about the same as that of a hydrogen maser. However, the second-order Doppler shift was relatively high and the S/N was relatively low, so this system was not developed further as a frequency standard.

A. 199Hg+ Paul Trap Frequency Standards

In 1973, Major and Werth observed the 40.5 GHz ground-

state hyperfine transition of 199 Hg $^+$ with a linewidth of a few Hz [36]. The Q of the resonance was approximately 10^{10} . 199 Hg $^+$ has some basic advantages as a frequency standard. Its hyperfine transition has a very high frequency. Because of its large mass, it has a low second-order Doppler shift at a given temperature.

The detection of the resonance is based on optical pumping. The lowest electronic levels are shown in Fig. 3. The ground electronic state of Hg⁺ has the electronic configuration $5d^{10}6s^2S_{1/2}$. An RF-excited lamp containing the 202 Hg isotope will emit 194 nm radiation that will drive 199 Hg⁺ ions in the F=1 hyperfine level of the ground state to the $5d^{10}6p^2P_{1/2}$ state. The ions can then decay to either the F=0 or F=1 hyperfine levels. The lamp eventually pumps most of the ions to the F=0 state. If microwave radiation near the 40.5-GHz resonance is applied, some ions are driven to the $m_F=0$ sublevel of the F=1 state. Then they can be excited to the $5d^{10}6p^2P_{1/2}$ state by light from the lamp. When they decay, the 194-nm photons are detected with a photomultiplier tube.

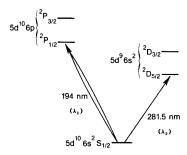


Fig. 3. Electronic energy levels of Hg⁺. The ground electronic state of the ¹⁹⁹Hg⁺ isotope is made up of two hyperfine levels, separated by 40.5 GHz. Some microwave frequency standards are based on this transition. An optical frequency standard might be based on the 281.5-nm transition.

Jardino et al. [5] made the first frequency standard based on this system. They measured $\sigma_y(\tau)=3.6\times 10^{-11}\tau^{-1/2}$, for $10<\tau<3500$, where τ is the time interval in seconds. This stability was comparable to that of some commercial cesium atomic clocks.

This basic system was developed further by Cutler et al. [6], [7], [24], [30]. They introduced helium buffer gas to reduce the temperature of the ions. The lamp was turned off when the microwave radiation was applied, in order to avoid shifts of the microwave resonance frequency. The number of ions was about 2×10^6 . The resonance linewidth was 0.85 Hz, so the Q was approximately 5×10^{10} . The results of a 115 day test showed fractional frequency fluctuations of 7.6×10^{-15} for integration times of 1 day [7]. A short term stability of $\sigma_y(\tau) < 2 \times 10^{-12} \tau^{-1/2}$ can be inferred from the published data [7]. The frequency difference between two standards was between one and two parts in 10^{-13} , which is an indication of their accuracy.

Prestage et al. [9] have demonstrated a 199Hg+ frequency standard based on a linear RF trap. Ramsey's separated oscillatory field method [37] was employed to drive the resonance. In this method, two short RF pulses are applied. This yields a linewidth in frequency (as opposed to angular frequency) units of about 1/(2T), where T is the time between the two pulses. This is about a factor of two narrower than is obtained by applying a single RF pulse of duration T. The frequency standard was operated with a linewidth of 0.16 Hz and a Q of 2.5×10^{11} . The shortterm stability of the device was $\sigma_y(\tau) = 1.6 \times 10^{-13} \tau^{-1/2}$ for $50 < \tau < 800$. Resonance linewidths as small as 0.03 Hz were observed when T was increased to 16 s. This corresponds to a Q of 1.3×10^{12} , the highest ever observed in a microwave atomic transition. A frequency standard based on a resonance line of this width could have a short term stability $\sigma_y(\tau) = 5 \times 10^{-14} \tau^{-1/2}$.

B. 9Be+ Penning Trap Frequency Standards

Bollinger et al. [10,11] demonstrated the first frequency standard based on laser-cooled ions. This standard was based on a 303-MHz hyperfine transition in the ground electronic state of ${}^9\mathrm{Be^+}$. The hyperfine sublevels of the ground state are shown in Fig. 4. The first derivative of the frequency of the transition between the $(m_I=-3/2,m_J=1/2)$ sublevel and the $(m_I=-1/2,m_J=1/2)$ sublevel approaches zero at a value of the magnetic field near 0.8194 T. These levels are labeled "1" and "2" in Fig. 4. A frequency-doubled cw dye laser was used to generate 313-nm radiation to laser-cool and optically detect the ions.

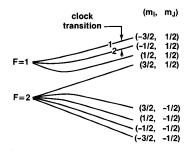


Fig. 4. ⁹Be⁺ energy levels in a magnetic field. The frequency standard is based on the 303 MHz "clock transition" between the levels labeled "1" and "2."

In the most recent version of the ${}^9\mathrm{Be}^+$ frequency standard, sympathetic laser cooling was used [12]. Magnesium ions were trapped at the same time and were continuously laser cooled. The number of ${}^9\mathrm{Be}^+$ ions was 5000 to 10 000. The 313-nm radiation source was tuned so that most of the ions would be pumped to the $(m_I=3/2,m_J=1/2)$ ground-state sublevel in a few seconds. The 313-nm radiation was turned off, and ions in the $(m_I=3/2,m_J=1/2)$ sublevel were transferred to the $(m_I=1/2,m_J=1/2)$ sublevel

and then to the $(m_I = -1/2, m_J = 1/2)$ sublevel by two successive RF pulses. The Ramsey method was then used to drive some of the ions to the $(m_I = -3/2, m_J = 1/2)$ sublevel. Then RF pulses were applied in the reverse order, to bring ions which had remained in the $(m_I =$ -1/2, $m_J = 1/2$) sublevel back to the $(m_I = 3/2, m_J =$ 1/2) sublevel. The 313-nm source was then turned back on and the fluorescence intensity was measured. The intensity was proportional to the $(m_I = 3/2, m_J = 1/2)$ population. If ions were left in the $(m_I = -3/2, m_J = 1/2)$ sublevel, there was a decrease in the intensity. The time between the two RF pulses was as long as 550 s, although 100 s was more typical. With T = 550 s, the width of the resonance was 900 μ Hz. The stability was better than $3\times 10^{-12}\tau^{-1/2}$ for $10^3<\tau<10^4.$ However, there was a frequency shift with changes in pressure. This limited the long-term stability of the standard to about 3×10^{-14} . The uncertainty of the second-order Doppler shift, though, was only 5×10^{-15} . The longest time that the standard was operated continuously was about ten hours.

D. Other Work

Other ions have been investigated for use in microwave frequency standards. Lasers have been used for optical pumping and detection of hyperfine transitions in several other ions, including 25 Mg⁺ [38], 137 Ba⁺ [39], 135 Ba⁺ [40], 171 Yb⁺ [41], and 173 Yb⁺ [42]. A frequency standard based on the 8-GHz hyperfine transition of 137 Ba⁺ has shown a stability comparable to that of some commercial cesium standards [13]. A resonance with a Q of 3.8×10^{11} has been observed in 171 Yb⁺, which has a 12.6-GHz hyperfine transition. A 171 Yb⁺ frequency standard has been tested, and a stability of $\sigma_y(50s) = 2 \times 10^{-12}$ has been reported [14].

In general, frequency standards based on Penning traps suffer from the fact that the transitions which are insensitive to magnetic field fluctuations at high magnetic fields have low frequencies. Frequency standards based on Paul traps suffer from having high second-order Doppler shifts, due to the micromotion. Wineland *et al.* proposed to use a linear RF trap to confine a single string of ions, such as ¹⁹⁹Hg⁺, along the central axis [15]. The ions could be laser cooled and would have negligible micromotion. Such a standard might combine high accuracy and high stability.

VI. OPTICAL FREQUENCY STANDARDS

An optical frequency standard might be based on a transition with a narrow natural linewidth. The Q could then be so high that the signal from even a single ion could yield good stability as well as good accuracy. The upper state of the transition must be metastable. Direct detection of the photons emitted by such a transition would be very difficult.

Sensitive detection of a single ion can be carried out by a double resonance method called electron shelving [18]. The Hg⁺ ion is an example of an atom that has a level structure that is suitable for this method (see Fig. 3). First, a pulse of resonant radiation is applied at wavelength λ_2 to

try to drive the transition to the metastable state. Then, in a time less than the lifetime of the metastable state, another pulse of radiation is applied at wavelength λ_1 . This radiation is resonant with a transition from the same lower state to a short-lived upper state. If the atom is shelved in the metastable state, no λ_1 photons are emitted from the short-lived state. If the atom is in the lower state after the λ_2 pulse, it can absorb and emit λ_1 photons at a high rate. Thus the absorption of a single λ_2 photon, which drives the atom to the metastable state, results in the absence of many λ_1 photons. Thus individual λ_2 transitions can be detected, even if not all of the λ_1 photons are detected.

A. Ba+ Single-Ion Optical Spectroscopy

The $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states of Ba⁺ are metastable. Janik *et al.* observed a Doppler-free two-photon resonance between the ground $6s^2S_{1/2}$ state and the $5d^2D_{3/2}$ state in a single Ba⁺ ion in a Paul trap [43]. The two-photon resonance was 3-MHz wide because of the laser linewidths. The transition has the potential of being much narrower than 1 Hz, because of the long lifetime of the $5d^2D_{3/2}$ state. Recently, the $6s^2S_{1/2}$ to $5d^2D_{5/2}$ one-photon transition has been observed in a single Ba⁺ ion, using electron shelving [17]. The width, which was limited by the laser, was about $50~\rm kHz$.

B. Hg+ Single-Ion Optical Spectroscopy

Hg⁺ has a level structure which is suitable for an optical frequency standard. The $5d^96s^2$ $^2D_{5/2}$ state is metastable, with a lifetime of about 90 ms. The 194-nm transition from the ground $5d^{10}6s^2S_{1/2}$ to the $5d^{10}6p^2P_{1/2}$ state can be used for laser cooling and for detection by electron shelving. Some hyperfine components of the 281.5-nm transition in ¹⁹⁹Hg⁺ are nearly independent of magnetic field, near zero field. One of these is the transition from F=0 in the ground state to $(F=2,m_F=0)$ in the upper state. Recently, Bergquist et al. [16] observed this transition with a linewidth of under 80 Hz. The resonance line Q is over 10¹³ and is the highest ever observed in an atomic transition. The laser frequency was servoed to the singleion resonance for periods of several minutes [16]. Further work on this system might yield a frequency standard with $\sigma_y(\tau) \approx 10^{-15} \tau^{-1/2}$ and accuracy of one part in 10¹⁸ [15].

C. Other Work

Many other ions, including Tl⁺, In⁺, Ga⁺, Al⁺, B⁺, Pb⁺, I⁺, and Bi⁺, have narrow optical transitions, and have been proposed as frequency standards [18], [44]. Experimental work is being carried out on Yb⁺ [14], [45] and Sr⁺ [46]. Both Yb⁺ and Sr⁺ have been trapped and laser-cooled, but narrow optical lines have not been observed yet.

VII. SUMMARY

Trapped-ion frequency standards are not yet in widespread operational use. The ¹⁹⁹Hg⁺ microwave frequency standard has demonstrated good stability. Other frequency standards based on laser-cooled ions are being developed and may be capable of better accuracy. Ultimately, the most accurate frequency standard may be based on an optical transition in a single, laser-cooled ion.

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Wayne M. Itano was born in Pasadena, CA in 1951. He received the B. S. degree in physics from Yale University, New Haven, CT in 1973 and the M. A. and Ph. D. degrees in physics from Harvard University, Cambridge, MA in

from Harvard University, Cambridge, MA in 1975 and 1979, respectively.

Since 1979, he has been a physicist at the National Institute of Standards and Technology in Boulder, CO. In 1979 and 1980 he was a National Research Council Postdoctoral Associate. In 1990 he was a Science and Technology Agency Fellow at the Communications Research Laboratory in Tokyo. He is currently Secretary-Treasurer of the Laser Science Topical Group of the American Physical Society. His research interests include the applications of laser cooling and high resolution spectroscopy of stored ions to basic

American Physical Society. His research interests include the applications of laser cooling and high resolution spectroscopy of stored ions to basic physics and to frequency standards.

Dr. Itano is a recipient of the Samuel Wesley Stratton Award from NIST, the Commerce Department Gold Medal, the David J. Robbins Prize from Harvard, and the De Forest Pioneers Prize from Yale. He is a member of Phi Beta Kappa, a fellow of the American Physical Society, and a member of the Ontical Society of America member of the Optical Society of America.